

## DESCRIPTION

METAL POWDER PRODUCTION PROCESS AND METAL COMPOUND FEED  
COMPACT

5

## TECHNICAL FIELD

The present invention relates to a metal powder production process and a metal compound feed compact (feed metal compound compact) that can be applied to this metal powder production process.

10

## BACKGROUND ART

High-melting-point rare metals such as niobium and tantalum not only have a high melting point, but since they are also chemically active at high temperatures, sophisticated technology is required for forming lump metals into powders. As one example of a process for producing this rare metal powder, an atomization method in which a metal is melted and is atomized the resulting metal liquid is one effective process for producing powder. However, an expensive equipment is required to melt rare metals because of their high melting point.

In addition, another process is known as the hydrogenation-dehydrogenation (HDH) method in which reacting a rare metal with hydrogen to form a hydride and mechanically crushing the resulting hydride. However, this process has difficulty in producing a fine powder of uniform particle size.

At present, powders of tantalum and other high-melting-point metals are produced industrially for use as materials of capacitors, and this powder is mainly produced by a metallothermic reduction method in which the raw material is reduced

using an active metal as the reducing agent. Use of a metallothermic reduction method offers the advantage of being able to produce powdered tantalum directly by a reduction reaction. However, it is difficult to efficiently produce a uniform powder in large volume by this reduction reaction, thereby resulting in the disadvantage of decreased  
5 uniformity of the resulting powder when the scale of the reaction is increased in particular.

Under the present circumstances, powdered tantalum is produced by using a large amount of halide-based molten salt for the reaction medium (dilute salt or dilluent), and adding small amounts of raw material and reducing agent to this reaction medium to  
10 enable a uniform reduction reaction. This process is suitable for the production of uniform powder at high purity. On the other hand, it has the shortcomings of requiring a large amount of the reaction medium salt, and difficulties in increasing the scale and speed of the process.

If the niobium is reduced by the process of using a large amount of reaction  
15 media salt by the method of the above-mentioned, a uniform powder is obtained. However, it is difficult to obtain a uniform powder when reducing it without using the reaction media salt as well as the production process of the tantalum.

#### DISCLOSURE OF THE INVENTION

20 The present invention provides a process for producing a metal powder by using a metal compound for the raw material and reducing said metal compound, comprising: a molding step in which the metal compound is mixed with a binder, is molded, and is sintered to produce a metal compound feed compact; and, a reducing step in which a metal is formed by reducing the metal compound feed compact by contacting the metal  
25 compound feed compact with an active metal as a reducing agent.

A niobium compound can be used for the metal compound. Alternatively, tantalum compound can be used for the metal compound. Alternatively, a compound of a metal element selected from zirconium, titanium, hafnium, rare earth metal and actinide metal can be used for the metal compound.

5 In the molding step, a metal compound feed compact can be molded by mixing a metal compound, a binder, and an active metal compound as a reaction agent.

At least one of active metal selected from calcium, magnesium, sodium, barium and potassium is preferably used for the reducing agent.

At least one of compound of an active metal selected from calcium, magnesium,  
10 sodium, barium and potassium is preferably used for the reaction agent.

A niobium oxide or niobium halide is preferably used for the niobium compound.

The temperature of the compact in the reducing step is preferably 600 to 1300°C.

15 In the molding step, the metal compound feed compact is preferably molded into a shape in which the distance from an arbitrary location within the metal compound feed compact to the surface of the compact is not longer than 10 mm.

The metal powder production process preferably additionally contains a step in which the metal formed in the reducing step is separated from the active metal and  
20 by-products by acid treatment.

In addition, the present invention provides a metal compound feed compact which is mixed a metal compound and a binder, is molded and is sintered; wherein the distance from an arbitrary location within the compact to the surface of the compact is not longer than 10 mm.

25 The metal compound preferably contains a compound raw material of a metal

element selected from niobium, zirconium, titanium, hafnium, tantalum, rare earth metal and actinide metal.

The metal compound feed compact may contain at least one of compound of a metal selected from calcium, magnesium, sodium, barium and potassium as a reaction agent. The reaction agent is preferably one selected from an oxide, halide, and carbonate of at least one of metal selected from calcium, magnesium, sodium, barium and potassium.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a cross-sectional block diagram showing a metal production device according to the present invention.

Fig. 2 is an electron microscopic photograph of niobium powders produced in Example 1.

Fig. 3 is an electron microscopic photograph of tantalum powders produced in Examples A through E of Example 2.

Fig. 4 is an electron microscopic photograph of tantalum powders produced in Examples F through K of Example 2.

Figs. 5A and 5B are graphs showing the particle size distributions of tantalum powders produced in Example 2.

#### BEST MODE FOR CARRYING OUT THE INVENTION

The metal powder production process according to the present invention is a process for producing a metal by using a metal compound as a raw material and reducing the metal compound, comprising a molding step in which the metal compound is mixed with a binder, is molded, and is sintered to produce a metal compound feed compact

(preform), and a reducing step in which a metal is formed by reducing the metal compound feed compact by contacting the metal compound feed compact with an active metal as a reducing agent. A niobium compound, tantalum compound, zirconium compound, titanium compound, hafnium compound, rare earth metal compound or  
5 actinide metal compound can be used for the metal compound.

As follows is more detailed description of a process for producing niobium powder using a niobium compound as an example of the metal powder production process according to the present invention.

The niobium powder production process according to the present invention is a  
10 process for producing niobium by using a niobium compound for the raw material and reducing said niobium compound, and comprises a molding step in which the niobium compound is mixed with a binder to produce a slurry and is molded this into a predetermined shape such as plates, wires or granules and is sintered to produce a niobium compound feed compact (preform), and a reducing step in which niobium is  
15 formed by reducing the niobium compound by contacting the niobium compound feed compact with an active metal as a reducing agent. A niobium oxide or niobium halide can be used for the aforementioned niobium compound.

At least one of metal selected from calcium, magnesium, sodium, barium and potassium is preferably used for the aforementioned active metal. The reduction  
20 reaction can be made to proceed efficiently by reducing the niobium compound feed compact using this type of active metal. Moreover, the use of magnesium for the active metal is particularly preferable since the reduction reaction can be made to proceed the most efficiently.

The temperature of the compact in the aforementioned reducing step is  
25 preferably 600 to 1300°C. If the compact temperature is below 600°C, the vapor

pressure of the reducing agent tends to be low and the reduction reaction tends to not proceed adequately, while if the compact temperature is above 1300°C, the sintering of the niobium powder formed tends to proceed.

The active metal is preferably contacted with the niobium compound feed compact (preform) in the vaporized state in the aforementioned reducing step. In this case, the niobium compound feed compact is preferably subjected to the reduction reaction by arranging in the reduction device so that ventilation is maintained. For example, by arranging niobium compound feed compacts in a vertical position using support materials in a reduction device having a heating device and support materials to support plural niobium compound feed compacts in the vertical position, arranging the active metal in the bottom of the reduction device, on the support materials or between adjacent niobium compound feed compacts, and heating the reduction device with the heating device, the active metal is vaporized and contacted with the niobium compound feed compacts thereby allowing the reduction reaction to take place. In this manner, by arranging niobium compound feed compacts in a reduction device so that diffusion path through gas phase is maintained, since the vaporized active metal is uniformly diffused, the niobium compound feed compacts and active metal can be allowed to make contact more uniformly, thereby enabling the reduction reaction to proceed more uniformly in terms of both time and space while also increasing reduction efficiency. In addition, since the niobium compound feed compacts do not make direct contact with the reduction device, contamination from the reduction device can be prevented, thereby increasing the purity of the resulting niobium powder.

In addition, in the aforementioned molding step for producing a niobium compound feed compact, the niobium compound feed compact can be produced by additionally mixing in a reaction agent an active metal compound as a reaction agent in

addition to the niobium compound and binder. Although an active metal compound as the aforementioned reaction agent can also be used as a binder, it can also be used to control the morphology of deposit of the niobium powder as well as increase acid treatment efficiency. In this manner, by producing the niobium compound feed compact by mixing in an active metal compound as a reaction agent in addition to the niobium compound and binder, the reaction in the reducing step proceeds more uniformly and the production efficiency can be increased. Moreover, the particle size of the resulting niobium powder can also be controlled by regulating the type and mixing ratio of the aforementioned reaction agent. In addition, contamination from the reaction vessel can also be effectively prevented.

A compound of at least one of metal selected from calcium, magnesium, sodium, barium and potassium is preferably used for the aforementioned reaction agent, and an oxide, halide, carbonate, hydroxide, chloride and/or fluoride of these metals is used more preferably. The reaction efficiency and uniformity of the reducing step can be further enhanced by using a compact containing this type of active metal compound as a reaction agent.

Moreover, in the aforementioned molding step, the shape of the niobium compound feed compact produced is preferably formed for the distance from an arbitrary location within the compact to the surface of the compact is not longer than 10 mm.

The aforementioned "distance from an arbitrary location within the compact to the surface of the compact" indicates the distance to the surface that is the shortest distance from an arbitrary location within the compact. In the reducing step of this production process, the niobium compound contained in the niobium compound feed compact is reduced as a result of the reducing agent diffusing from the surface of the niobium compound feed compact to the inside of the compact. Thus, if the shape of the compact

is that described above, the maximum diffusion distance of the reducing agent from the surface of the compact at completion of the reduction reaction is not longer than 10 mm, diffusion treatment of the reducing agent that diffuses from the surface of the compact to inside the compact does not vary significantly according to the site, and the niobium raw material can be uniformly and rapidly reduced throughout the compact.

Moreover, the aforementioned niobium powder production process may additionally contain a step in which the niobium formed in the aforementioned reducing step, reducing agent, reaction agent and by-products are separated by acid treatment. By providing a step in which these substances are separated by acid treatment, the reducing agent, reaction agent and by-products formed after the reduction reaction can easily be separated from the objective niobium, enabling the large-volume production of high-purity niobium. In addition, in the production process according to the present invention, since a niobium compound feed compact is used in the reaction, the shape of the aforementioned compact is nearly maintained even after reduction. Consequently, by performing acid treatment on the compact following this reduction, there is the advantage of being able to efficiently remove the reducing agent and by-products.

In conventional rare metal production processes, it is considered that the reason for the the greatest not to obtain a uniform powder is that no uniformity is caused by the part of raw material in the progression condition of the reduction reaction timewise and spatially since the diffusion of the reducing agent in the raw material becomes a ununiformity when the amount of reduction is increased. In contrast, in the niobium powder production process according to the present invention, since a niobium compound feed compact is produced by mixing niobium raw material, binder and/or reaction agent, and a reduction reaction is carried out by contacting this mixture with an active metal, the reduction reaction is able to proceed uniformly in terms of both time



and space even if the amount of reduction increases. In addition, since the reduction reaction is allowed to proceed by placing the niobium compound feed compact in a reduction device, continuous operation and increased scale of the reduction process as well as improved efficiency of batch processing can be carried out with a simple device.

5 Namely, although continuous operation and acceleration of the process were difficult with the conventional metallothermic reduction reaction, since the niobium compound feed compact used in the aforementioned niobium powder production process makes it possible easily achieve a continuous work flow with respect to production, heat treatment, reduction, cleaning and so forth, the process can easily be operated continuously and  
10 increased in scale. In addition, although there were many cases in which the particle size and so forth of the resulting powder was not uniform and productivity was low if the amount of raw material was increased in the processes of the conventional method, according to the aforementioned production process, increased scale and speed of the reduction process can easily be achieved while maintaining the uniformity of the particle  
15 size of the resulting powder, thereby making it possible to improve productivity.

Next, the present invention will be explained in more detail, referring the figures of a first embodiment of the present invention. Fig. 1 is a cross-sectional view showing one embodiment of a metal production device according to the present invention.

The production device shown in Fig. 1 is composed of a reaction vessel 10  
20 which is a sealed vessel composed of a heat-resistant material such as stainless steel, a plurality of plate-like metal compound feed compacts 12 installed in reaction vessel 10, and support materials 13 and 14 provided above and below compacts 12 for supporting these compacts 12 in the vertical position. An active metal 15 which is a reducing agent reducing the metal raw material of compacts 12 is introduced into the bottom of reaction  
25 vessel 10. Furthermore, although a heating unit is not shown in the reaction vessel 10

shown in the figure, a heating unit can be provided for heating the inside of reaction vessel 10 to a predetermined temperature and vaporizing active metal 15.

The metal compound contained in compacts 12 is reduced to form a metal by diffusing a gas of active metal 15 within reaction vessel 10 by heating and vaporizing active metal 15 with the aforementioned heating unit, and then diffusing active metal 15 which is a reducing agent from the surface to the inside of compacts 12.

In the niobium powder production process according to the present invention, niobium powder can be produced by a metallothermic reduction method that uses the aforementioned production device. In order to produce the aforementioned niobium powder, a niobium compound feed compact containing a niobium compound which is the raw material is first produced in a molding step. This compact is obtained by mixing this niobium raw material with a binder and reaction agent, molding to a predetermined shape, and then firing at a temperature of preferably 300 to 1000°C, and more preferably 800 to 1000°C, to remove the binder. If the aforementioned firing temperature is lower than 300°C, since the binder and reaction agent are unable to be adequately removed, the reaction efficiency in the following reducing step tends to decrease, and the purity of the resulting powder also tends to decrease. If the firing temperature exceeds 1000°C, sintering proceeds excessively rapidly, which tends to cause a considerable change in the niobium compound feed compacts. In addition, although dependent on the dimensions and heating temperature of the compacts, this firing step is preferably carried out for 1 to 12 hours, more preferably for 1 to 6 hours, and even more preferably for 3 to 6 hours. In addition, in the case of carrying out firing, it is preferable to carry out in air or an oxygen atmosphere.

Examples of niobium compounds that can be used for the niobium raw material include  $\text{Nb}_2\text{O}_5$ ,  $\text{NbO}_x$  (preferably a niobium lower oxide wherein X is 0.5 to 2.5),  $\text{NbCl}_5$

and  $K_2NbF_7$ . The mixing ratio of niobium raw material in the aforementioned niobium compound feed compacts is preferably not less than 10% by weight, and more preferably not less than 50% by weight. Although the reduction reaction is carried out adequately if the mixing ratio of the niobium raw material is not less than 10% by weight, if the  
5 mixing ratio of the niobium raw material is less than 50% by weight, the amount of acid used increases during separation of niobium powder by acid treatment, which tends to cause a decrease in leaching efficiency.

Any binder can be applied without problem in the case that it can be easily removed by heat treatment regardless of the reduction reaction, examples of which that  
10 can be used include collodion and cellulose. In addition, a reaction agent can also be used as a binder, and a compact can be formed by mixing a niobium raw material and reaction agent. The mixing ratio of binder in the aforementioned niobium compound feed compact is preferably 5 to 80% by weight, more preferably 30 to 70% by weight, and even more preferably 30 to 50% by weight. If the mixing ratio of binder is less  
15 than 5% by weight or more than 80% by weight, the strength of the compact tends to decrease and it tends to be difficult to form the compact.

The reaction agent is added to the niobium raw material and binder as necessary to form a compact by them. The reaction efficiency in the reducing step can be enhanced and the reduction of the niobium raw material can be carried out more  
20 uniformly by dispersing the reaction agent in the compact. A compound of at least one of metal selected from the group consisting of calcium, magnesium, sodium, barium and potassium is preferably used for this reaction agent, and oxides, halides, carbonates, hydroxides, chlorides and/or fluorides of these metals are used more preferably. More specifically, calcium chloride, calcium carbonate, sodium carbonate or sodium chloride  
25 and so forth can be used alone or as a mixture. The mixing ratio of the aforementioned

reaction agent is preferably such that cations in the reaction agent are preferably blended at 0 to 2 moles and more preferably at 0.5 to 1 mole with respect to 1 mole of niobium contained in the compact. In the case the cation in the reaction agent exceeds 2 moles with respect to 1 mole of niobium contained in the compact, the amount of acid used for acid washing tends to increase and leaching efficiency tends to decrease when separating the formed niobium by acid treatment.

In addition, the shape of the compact produced in the aforementioned molding step is such that the shortest distance from an arbitrary location within the compact to the surface of the compact is preferably not longer than 10 mm and more preferably 2 to 5 mm. As a result of having this shape, since the diffusion distance of the reducing agent that diffuses from the surface of the compact towards the inside of the compact in the reducing step to be described later can be made to be not longer than 10 mm, the reduction reaction can be carried out rapidly and uniformly. Examples of specific shapes include plates, wires or granules. In the case of having the shape of a plate, the plate thickness is preferably not thicker than 20 mm in order to satisfy the aforementioned conditions, and in the case of having the shape of a wire, the distance between the outer periphery and the center in a cross-section that is perpendicular to the center line in the direction of length is preferably not longer than 10 mm. In the case of having the shape of granules, the diameter is preferably not longer than 20 mm.

Next, a compact obtained according to the aforementioned molding step is introduced into a reaction vessel 10 as shown in Fig. 1. In Fig. 1, the compacts 12 are plates or wires, and although they are shown in the case of being supported in the vertical position, the manner in which compacts 12 are arranged in the vessel is not limited to the arrangement shown in the figures, but rather their arrangement can be suitably changed according to the shape of the compacts.

In addition, an active metal as a reducing agent 15 is introduced into the bottom of reaction vessel 10. One or two or more of metal selected from the group consisting of calcium, magnesium, sodium, barium and potassium is preferably used for this active metal 15. In the production device shown in the figures, although active metal 15 is arranged in the bottom of the vessel, active metal 15 may also be arranged on, for example, support material 14 or arranged between compacts 12 supported in the vertical position. The aforementioned active metal 15 is preferably introduced at 50 to 400 parts by weight and more preferably at 100 to 300 parts by weight with respect to 100 parts by weight of the aforementioned compacts 12 within reaction vessel 10. In the case of using the aforementioned active metal 15 at less than 50 parts by weight, the reduction reaction tends to not proceed adequately, and even if used at more than 400 parts by weight, the reduction efficiency tends not to improve.

Next, preparations for the reducing step are completed by sealing reaction vessel 10.

Compacts 12 and active metal 15 are introduced into reaction vessel 10, and this reaction vessel 10 is then arranged in a heating furnace and heated. Active metal 15 is vaporized by heating and together with filling the inside of vessel 10, and it is contacted with compacts 12. The reduction reaction proceeds starting on the surface of compacts 12, the niobium raw material contained in compacts 12 is reduced and metal niobium is formed.

In this reducing step, compacts 12 are preferably heated to a temperature of 600 to 1300°C and more preferably 800 to 1000°C. In the case the aforementioned temperature is lower than 600°C, the vapor pressure of active metal 15 as the reducing agent tends to be low and the reduction reaction tends not to proceed adequately, while in the case the temperature is higher than 1300°C, sintering of the niobium powder formed

tends to proceed. Although depending on the dimensions of compacts 12, heating temperature and so forth, the reaction time is roughly 1 to 6 hours, and more specifically, the reduction reaction is completed adequately if the reaction time is 1 hour or more in the case of the compacts 12 being in the shape of plates having a thickness of several millimeters and heating at a temperature of 800°C or higher.

Next, the compacts for which the reduction reaction has been completed are removed from vessel 10 after allowing reaction vessel 10 to cool. The shape of compact 12 before the reaction remains almost unchanged in the shape of the compact after the reaction, it is composed almost by the generated niobium, the compound of the active metal generated by reduction (by-products), the reducing agent of excess, and the reaction agent. Next, the formed niobium can be separated from the active metal compounds, reducing agent, reaction agent and reaction products that formed due to reduction by performing acid treatment on the compacts removed from the reaction vessel, thereby allowing the obtaining of niobium powder. This acid treatment is carried out by, for example, acid substitution treatment using water or organic solvent after performing acid leaching. Various acids such as hydrochloric acid, acetic acid, nitric acid, hydrofluoric acid or sulfuric acid can be used for the aforementioned acid leaching.

In the production process according to the present invention, since compacts containing formed niobium are obtained that have nearly the same shape as initially loaded compacts 12 following completion of a reduction reaction, acid permeability with respect to acid treatment in a subsequent separation step is satisfactory, enabling the niobium powder to be separated rapidly and uniformly.

In this manner, a compound feed compact according to the present invention is possible to easily achieve a continuous work flow with respect to a heat treatment

(molding) step, reduction step and acid treatment step and so forth in its production process, easily accommodate increases in compact size and quantity in cases of large-volume processing, and prevent impairment of reaction rate and reaction uniformity even if the number of compacts processed is increased. Thus, according to the  
5 production process and compound feed compact according to the present invention, the process can easily be operated continuously and increased in scale.

In addition, according to the niobium powder production process according to the present invention, since reduction of a raw material can be carried out rapidly and uniformly by carrying out a reduction reaction mediated by a compact that contains a  
10 niobium raw material, it is effective in that the resulting niobium powder has high purity and uniform particle size.

Although a detailed explanation of a niobium powder production process was provided in the aforementioned example, the production process according to the present invention can similarly also be applied to the production of other metal powders.  
15 Namely, while using a compound of a target metal as the raw material, after mixing this with a binder and molding to a predetermined shape in the molding step followed by firing to produce a metal compound feed compact, the resulting metal compound feed compact is reduced by an active metal in a reducing step, followed preferably by separating the active metal and by-products by acid treatment to produce the target metal  
20 powder. Compounds containing metal elements such as zirconium, titanium, hafnium, tantalum, rare earth metals or actinide metals are preferably used for the aforementioned metal compounds.

Next, a tantalum powder production process according to the present invention is explained in more detail. The aforementioned tantalum powder production process is  
25 a process for producing tantalum by using a tantalum compound as a raw material and

reducing said tantalum compound, and is comprised of a molding step in which the  
aforementioned tantalum compound is mixed with a binder to produce a slurry, molded  
this into a predetermined shape such as plates, wires or granules and sintered to produce  
a tantalum compound feed compact, and a reducing step in which tantalum is formed by  
5 reducing the aforementioned tantalum compound by contacting an active metal as a  
reducing agent with the aforementioned tantalum compound feed compact. Examples  
of tantalum compounds that can be used include tantalum oxides and tantalum halides.

The firing temperature in the aforementioned molding step is preferably 300 to  
1000°C and more preferably 800 to 1000°C, and the firing time, although dependent on  
10 the dimensions of the compact, heating temperature and so forth, is preferably about 0.5  
to 12 hours, more preferably about 1 to 6 hours and even more preferably about 1 to 3  
hours. If the aforementioned firing temperature is lower than 300°C, the binder is  
unable to be adequately removed from the compact, which tends to cause a decrease in  
the reaction efficiency in the subsequent reducing step as well as a decrease in the purity  
15 of the resulting powder, while if the temperature exceeds 1000°C, the tantalum  
compound feed compact tends to be deformed. In addition, in the case of carrying out  
firing, it is preferably carried out in air or in an oxygen atmosphere.

At least one of metal selected from the group consisting of calcium, magnesium,  
sodium, barium and potassium is preferably used for the aforementioned active metal.

20 The reduction reaction can be made to proceed efficiently by reducing the tantalum  
compound feed compact using this type of active metal. Moreover, the use of  
magnesium for the active metal is particularly preferable since the reduction reaction can  
be made to proceed the most efficiently.

The temperature of the compact in the aforementioned reducing step is  
25 preferably 600 to 1300°C and more preferably 800 to 1000°C. If the compact



temperature is below 600°C, the vapor pressure of the reducing agent tends to be low and the reduction reaction tends to not proceed adequately, while if the compact temperature is above 1300°C, the sintering of the tantalum powder formed tends to proceed. In addition, although depending on the dimensions of the tantalum compound feed compact, heating temperature and so forth, the reaction time in the aforementioned reducing step is about 1 to 24 hours, preferably about 1 to 6 hours and more preferably about 3 to 6 hours.

In addition, the active metal is preferably contacted with the tantalum compound feed compact in the vaporized state in the aforementioned reducing step. In this case, the tantalum compound feed compact is preferably subjected to the reduction reaction by arranging in the reduction device so that ventilation is maintained. For example, by arranging tantalum compound feed compacts in a vertical position using support materials in a reduction device having a heating device and support materials for supporting a plurality of tantalum compound feed compacts in the vertical position, arranging the active metal in the bottom of the reduction device, on the support materials or between adjacent tantalum compound feed compacts, and heating the reduction device with the heating device, the active metal is vaporized and contacted with the tantalum compound feed compacts thereby allowing the reduction reaction to take place. In this manner, by arranging tantalum compound feed compacts in a reduction device so that ventilation is maintained, since the vaporized active metal is uniformly diffused, the tantalum compound feed compacts and active metal can be allowed to make contact more uniformly, thereby enabling the reduction reaction to proceed more uniformly in terms of both time and space while also increasing reduction efficiency. In addition, since the tantalum compound feed compacts do not make direct contact with the reduction device, contamination from the reduction device can be prevented, thereby enhancing the purity of the resulting tantalum powder.

In addition, in the aforementioned molding step for producing a tantalum compound feed compact, the tantalum compound feed compact can be produced by additionally mixing in an active metal compound as a reaction agent in addition to the tantalum compound and binder. Although an active metal compound the  
5   aforementioned reaction agent can also be used as a binder, it can also be used to control the morphology of deposit of the tantalum powder as well as increase acid treatment efficiency. In this manner, by producing the tantalum compound feed compact by mixing in an active metal compound as a reaction agent in addition to the tantalum compound and binder, the reaction in the reducing step proceeds more uniformly and the  
10   production efficiency can be increased. Moreover, the particle size of the resulting tantalum powder can also be controlled by regulating the type and mixing ratio of the aforementioned reaction agent. In addition, contamination from the reaction vessel can also be effectively prevented.

A compound of at least one of metal selected from calcium, magnesium, sodium,  
15   barium and potassium is preferably used for the aforementioned reaction agent, and an oxide, halide, carbonate, hydroxide, chloride and/or fluoride of these metals is used more preferably. More specifically, calcium chloride, calcium carbonate, sodium carbonate or sodium chloride and so forth can be used alone or as a mixture. The mixing ratio of the aforementioned reaction agent is preferably such that cations in the reaction agent are  
20   preferably blended at 0 to 2 moles and more preferably at 0.5 to 1 mole with respect to 1 mole of tantalum contained in the compact. The reaction efficiency and uniformity of the reducing step can be further enhanced by using a compact containing this type of active metal compound as a reaction agent.

Moreover, in the aforementioned molding step, the shape of the tantalum  
25   compound feed compact produced is preferably molded such that the distance from an

arbitrary location within the compact to the surface of the compact is not longer than 10 mm. The aforementioned "distance from an arbitrary location within the compact to the surface of the compact" indicates the distance to the surface that is the shortest distance from an arbitrary location within the compact. In the reducing step of this production process, the tantalum compound contained in the compact is reduced as a result of the reducing agent diffusing from the surface of the tantalum compound feed compact to the inside of the compact. Thus, if the shape of the compact is that described above, the maximum diffusion distance of the reducing agent from the surface of the compact at completion of the reduction reaction is not longer than 10 mm, diffusion treatment of the reducing agent that diffuses from the surface of the compact to inside the compact does not vary significantly according to the site, and the tantalum raw material can be uniformly and rapidly reduced throughout the compact.

Moreover, the aforementioned tantalum powder production process may additionally contain a step in which the tantalum formed in the aforementioned reducing step, reducing agent, reaction agent and by-products are separated by acid treatment. By providing a step in which these substances are separated by acid treatment, the target tantalum, reducing agent, reaction agent and by-products formed accompanying the reduction reaction can easily be separated, enabling the large-scale production of high-purity tantalum. In addition, in the production process according to the present invention, since a tantalum compound feed compact is used in the reaction, the shape of the aforementioned compact is nearly maintained even after reduction. Consequently, by performing acid treatment on the compact following this reduction, there is the advantage of being able to efficiently remove the reducing agent and by-products.

Although the present invention is explained by the examples in more detail, the following examples are not intended to limit the present invention.

#### Example 1

5 In the present example, niobium was produced by a metallothermic reduction reaction using the production device shown in Fig. 1.

Reaction agent (selected from one or two or more of  $\text{CaCl}_2$ ,  $\text{CaCO}_3$ ,  $\text{Na}_2\text{CO}_3$  and  $\text{NaCl}$ ) was added at 0 to 80% by weight to a niobium raw material ( $\text{Nb}_2\text{O}_5$ , 100 g).

More specifically, the amount of reaction agent added was made to be such that the  
10 amount of cation ( $\text{Ca}^{2+}$ ,  $\text{Na}^+$ ) was 0, 1/10, 1/5 or 1/2 mole with respect to 1 mole of Nb.

Next, the aforementioned niobium raw material and reaction agent were mixed with a binder to produce slurry. 5% collodion solution (5% nitrocellulose, 40% ether and 55% ethanol) was used for the binder in a volume equal to the aforementioned raw material and reducing agent.

15 After adding a suitable amount of acetone to the resulting slurry to adjust the viscosity, the slurry was poured into a cast having of several millimeters thick and 20 cm square to produce a plurality of compacts. The compacts were prepared in two thicknesses of about 3 mm and about 6 mm.

Next, the resulting compacts were sintered for 1 hour in a furnace having an air  
20 atmosphere at  $1000^\circ\text{C}$  to completely remove the binder, solvent and moisture from the mixture of niobium raw material and reaction agent. As a result of carrying out this firing, the reducing agent was able to be conserved and carbon contamination of the formed niobium powder was able to be prevented.

A plurality of the resulting compacts were placed in a reaction vessel 10 along  
25 with a magnesium reducing agent (active metal 15) while ensuring ventilation and

sealing reaction vessel 10. Next, reaction vessel 10 containing compacts 12 and reducing agent 15 was placed in an electric furnace held at 1000°C for 6 hours, and after allowing the reduction reaction to proceed adequately, the entire vessel was removed from the furnace and allowed to cool.

5           After rough washing the resulting compacts following reduction (mixture of metal niobium powder, auxiliary agent, MgO and Mg) with aqueous acetic acid solution (1+1), the compacts were washed twice with 1 N aqueous HCl solution followed by substituting with water, alcohol and acetone and then drying.

          Evaluation of resulting niobium: Fig. 2 shows photograph of the state of the  
10   resulting niobium powder by a scanning electron microscope after reducing the niobium raw material by reacting the compound feed compacts in 50 x 20 x 3 mm square with magnesium vapor for 6 hours at 1000°C, allowing to cool, rough washing with aqueous acetic acid solution (1+1), washing twice with 1 N aqueous HCl solution, substituting with water, alcohol and acetone and drying. Furthermore, the photographs shown in Fig.  
15   2 are arranged in a matrix according to the type of auxiliary agent (horizontal axis) and cation ratio (vertical axis).

          As shown in this figures, the particle size of the niobium powder was found to be able to be controlled by changing the type and amount of the auxiliary agent in the compound feed compacts. The horizontal width of one of the photographs is about 15  
20   μm, and although the powder size depends on the type and amount of auxiliary agent, niobium powder was obtained having a primary particle size of about 0.5 to 3 μm.

## Example 2

          In the present example as well, tantalum was produced by a metallothermic  
25   reduction reaction using the production device shown in Fig. 1.

Slurries were prepared by mixing tantalum raw material ( $\text{Ta}_2\text{O}_5$ ), reaction agent (selected from at least one of  $\text{CaCl}_2$ ,  $\text{CaCO}_3$ ,  $\text{Na}_2\text{CO}_3$  and  $\text{NaCl}$ ) and binder in the composition shown in Table 1. 5% collodion solution (5% nitrocellulose, 40% ether and 55% ethanol) was used for the binder in a volume equal to the aforementioned  
5 tantalum raw material and reaction agent. The viscosity of the aforementioned slurries was adjusted by changing the amounts of reaction agent and binder added.

The resulting slurries were then poured into a cast to produce plate-shaped compacts having a thickness of 5 to 10 mm.

Next, the resulting compacts were sintered for 3 hours in a furnace having an air  
10 atmosphere at  $1000^\circ\text{C}$  to completely remove the binder, solvent and moisture from the mixture of tantalum raw material and reaction agent. As a result of carrying out this firing, the reducing agent was able to be conserved and carbon contamination of the formed tantalum powder was able to be prevented.

4 to 10 sheets of the resulting compacts were placed in a reaction vessel 10  
15 along with 20 g of magnesium reducing agent (active metal 15) while ensuring ventilation and were sealed reaction vessel 10 welded by tungsten inert gas. Next, reaction vessel 10 containing compacts 12 and reducing agent 15 was placed in an electric furnace held at  $700$  to  $1000^\circ\text{C}$  for 6 to 24 hours, and after allowing the reduction reaction to proceed adequately, the entire vessel was removed from the furnace and  
20 allowed to cool.

After rough washing the resulting compacts following reduction (mixture of metal tantalum powder, auxiliary agent,  $\text{MgO}$  and  $\text{Mg}$ ) with aqueous acetic acid solution (1+1), the compacts were washed twice with 1 N aqueous  $\text{HCl}$  solution followed by substituting with water, alcohol and acetone and then drying.

Table 1

Example	Type of reaction agent	X	Mixing ratio		Reducing step	
			Ta <sub>2</sub> O <sub>5</sub> (g)	Reaction agent (g)	Time (hr)	Temp. (°C)
A	CaCl <sub>2</sub>	1	5.18	2.61	6	1000
B	CaCO <sub>3</sub>	1	4.96	2.25	6	1000
C	CaO	1	5.16	1.29	6	1000
D	Na <sub>2</sub> CO <sub>3</sub>	1	5.00	1.15	6	1000
E	NaCl	1	5.03	1.40	6	1000
F	--	0	5.07	--	6	1000
G	CaCl <sub>2</sub>	0.1	5.02	0.27	6	1000
H	CaCl <sub>2</sub>	0.2	5.03	0.50	6	1000
I	CaCl <sub>2</sub>	0.5	5.12	1.26	6	1000
J	CaCl <sub>2</sub>	1	5.10	2.52	6	1000
K	CaCl <sub>2</sub>	2	5.02	4.98	6	1000
L	CaCl <sub>2</sub>	1	5.01	2.54	6	700
M	CaCl <sub>2</sub>	1	5.01	2.54	6	800
N	CaCl <sub>2</sub>	1	5.01	2.54	6	900
O	CaCl <sub>2</sub>	1	5.01	2.54	6	1000
P	CaCl <sub>2</sub>	1	3.05	1.52	3	1000
Q	CaCl <sub>2</sub>	1	3.04	1.52	6	1000
R	CaCl <sub>2</sub>	1	3.03	1.56	12	1000
S	CaCl <sub>2</sub>	1	3.04	1.55	24	1000

Furthermore, in Table 1, X represents the number of moles of cation contained in the reaction agent with respect to 1 mole of tantalum.

5 The states of the resulting tantalum powders were observed using a scanning electron microscope, and particle size distribution was investigated by optical diffraction analysis.

Figs. 3 and 4 are photographs showing the states of the tantalum powders obtained in Examples A through K by a scanning electron microscope. As shown in Fig. 10 3, the resulting tantalum powders were in the form of coral-like particles having a particle size of 0.1 to 0.5  $\mu\text{m}$ . In addition, in the case of using a calcium compound for the reaction agent, the particle size of the resulting tantalum powders was uniform. In contrast, in the case of using NaCl or NaCO<sub>3</sub> for the reaction agent, the resulting

tantalum powder contained two types of particles consisting of those having a small particle size of 0.1 to 0.2  $\mu\text{m}$  and those having a large particle size of about 0.5  $\mu\text{m}$ . In addition, as shown in Fig. 4, the particle size of the resulting tantalum powder by increasing the mixing ratio of reaction agent in the compacts.

5        Fig. 5A is a graph showing the particle size distribution of tantalum powders obtained in Examples B, D and O. As is indicated in the graph, in the case of using a sodium compound for the reaction agent, powders having a smaller particle size tend to be obtained more than in the case of using a calcium compound. In addition, it was also shown that in the case of using a calcium compound for the reaction agent, the particle  
10      size of the resulting tantalum powder was more uniform.

Fig. 5B is a graph showing the particle size distribution of tantalum powders obtained in Examples H, I and O. As is indicated in this graph, it was shown that the particle size of the resulting tantalum powder increases as a result of increasing the mixing ratio of reaction agent in the compact.

15        On the basis of these results, it was shown that the particle size of tantalum powders can be controlled by changing the type and amount of reaction agent in the compound feed compact.

## INDUSTRIAL APPLICABILITY

20        As is explained in detail above, according to the niobium powder production process according to the present invention, as a result of allowing a reduction reaction to proceed uniformly as well as allowing a reduction reaction to proceed by placing a niobium compound feed compact in a reduction device, the reduction process can be operated continuously, the scale of the process can be increased and batch processing can  
25      be carried out efficiently with a simple device, thereby making it possible to efficiently



produce high-purity niobium powder directly from a niobium compound.

In addition, according to the tantalum powder production process of the present invention, as a result of allowing a reduction reaction to proceed uniformly as well as allowing a reduction reaction to proceed by placing a tantalum compound feed compact in a reduction device, the reduction process can be operated continuously, the scale of the process can be increased and batch processing can be carried out efficiently with a simple device, thereby making it possible to efficiently produce high-purity tantalum powder directly from a tantalum compound.

In addition, according to the metal powder production process of the present invention, a powder of a metal other than niobium and tantalum such as zirconium, titanium, hafnium, rare earth metals and actinide metals can be produced efficiently at high purity from a metal compound, and preferably can be produced continuously.

In addition, a metal compound feed compact according to the present invention is a compact of a metal compound comprised by mixing the metal compound with a binder and then molding to a predetermined shape, firing, and as a result of being provided with a constitution in which the distance from an arbitrary location within the aforementioned compact to the surface of the compact is not longer than 10 mm, the diffusion distance of a reducing agent from the surface of the compact can be made to be not longer than 10 mm, thereby enabling uniform and rapid reduction of the metal compound.